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PROVISIONAL SPECIFICATION

Melt Blending of Aromatic Polyesters with other Polymeric Materials

We, JAMES TENNANT DICKSON AND REGINALD JOHN WILLIAM REYNOLDS, both of Hexagon House, Blackley, Manchester, British Subjects, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1., a Company incorporated under the laws of Great Britain, do hereby declare the nature of this invention to be as follows:

10 This invention relates to the melt blending of aromatic polyesters with other polymeric materials and more particularly with other linear condensation polymers.

15 The aromatic polyesters with which the present invention is concerned are the highly polymeric linear esters obtainable by heating glycols of the series $\text{HO}(\text{CH}_2)_n\text{OH}$, where n is an integer greater than 1 but not exceeding 10, with terephthalic acid or with an ester-forming derivative thereof, for example, an aliphatic (including cycloaliphatic) or aryl ester or half-ester, an acid halide or an ammonium or amine salt, under conditions which yield the esters in a highly polymerised condition. Examples of such highly polymeric linear esters are those obtainable from terephthalic acid or an ester-forming derivative thereof and ethylene glycol, trimethylene glycol, tetramethylene glycol, hexamethylene glycol and decamethylene glycol. These polyesters are high melting, difficultly soluble, colourless or substantially colourless materials which can be formed into filaments which can be extended by drawing into strong flexible fibres showing, by characteristic X-ray patterns, molecular orientation along the fibre axis. Of these polyesters, polyethylene terephthalate is preferred on account of its ready availability and of its outstanding utility as a textile material.

We have now found that these aromatic polyesters may be satisfactorily blended by melting with a wide variety of other

synthetic linear condensation polymers.

According to the present invention we provide blends of aromatic polyesters of the kind hereinbefore specified with other synthetic linear condensation polymers, said blends being obtained by heating said polyesters and the other polymers together above the melting points thereof at least until a homogeneous blend is obtained.

Synthetic linear condensation polymers which may be blended in accordance with the present invention include polyamides and interpolyamides of all kinds, for example, polyhexamethylene adipamide, polyhexamethylene sebacamide, the polyamides obtainable from caprolactam, the interpolyamides obtainable from caprolactam and hexamethylenediammonium adipate and the interpolyamides from hexamethylenediammonium adipate, hexamethylenediammonium sebacate and caprolactam; polyesters and interpolyesters (including different aromatic polyesters from the class hereinbefore specified), for example, polytrimethylene terephthalate, polyhexamethylene terephthalate polydiethyleneglycol terephthalate, polyethylene-1:2-ethane bis(phenoxy-p-carboxylate), polyethylene adipate, polyethylene sebacate and polyethylene hexahydroterephthalate; polyurethanes and polyureas.

In carrying the invention into practical effect the components are mixed together and heated at a temperature at least sufficiently elevated to cause the components to melt, and the heating is continued until a homogeneous mass is obtained. Blending may be facilitated by stirring and if desired by the use of reduced pressures. Advantageously, the melting is effected in the absence of air or oxygen and, if desired, a stream of an oxygen-free gas, for example, nitrogen, may be passed through the melts to assist in the formation of a homogeneous

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blend. If desired, the blending operation can be preceded by a drying operation.

The melts obtained at the conclusion of the heating operation may be extruded or otherwise removed from the vessel and subsequently cooled. The extruded melts may be formed into blocks, chips or other shapes.

10 The blends obtained according to the present invention usually find utility in the art in the form of filaments or fibres. Filaments can be formed by extrusion
15 by drawing from the melt directly after the completion of the heating or after re-melting. Any suitable apparatus may be used. The filaments can be subsequently drawn to as much as several times their original lengths. The drawing operation may be carried out on
20 filaments which have been allowed fully to cool and solidify after their formation, or it may directly follow the formation of the filaments as one part of a continuous process. Any suitable apparatus and process may be used for the drawing. For instance, the filaments may be wound from one roller to another, the second roller rotating at a higher speed
30 than the first roller, for example at a speed up to about 4 or 5 times that of the first roller.

The invention is illustrated but not limited by the following Examples, in
35 which the parts are expressed by weight.

EXAMPLE 1

18 parts of polyethylene terephthalate and 2 parts of polyhexamethylene adipamide, both in the form of small chips,
40 are mixed together in a suitable vessel, dried by heating at 200°C. at a pressure of 15 mm. of mercury for 3 hours and heated at 275°C. in a current of oxygen-free nitrogen at the same pressure. The

mass melts and becomes homogeneous after 3 hours heating. The blend is then extruded from the vessel in which it has been made and cooled. It is crystalline, has a melting point of 225°C.,
45 and it can be formed from the melt into filaments which can be extended by drawing into useful fibres.

EXAMPLE 2

Equal parts of polyethylene terephthalate and the interpolyamide formed from hexamethylenediammonium adipate, hexamethylenediammonium sebacate and caprolactam (40:30:30), both in the form of small chips, are mixed together
55 in a suitable vessel, which is fitted with a stirrer, and dried by heating for 3 hours at 200°C. at a pressure of about 15 mm. of mercury in a current of oxygen-free nitrogen. The interpolyamide melts during this heating. The temperature is increased to 275°C. and the heating is continued otherwise under the same conditions. The polyethylene terephthalate melts and after about 1½
60 hours a homogeneous blend is obtained. The blend is then extruded from the vessel and cooled. It is crystalline, has a melting point of 230°C., and it can be formed from the melt into filaments which can be extended by drawing into useful fibres.

EXAMPLE 3

The recipe of Example 2 is repeated except that an equal part of polyethylene adipate is used in place of the interpolyamide. The blend obtained is crystalline, melts at 148°C., and it can be formed from the melt into filaments which are somewhat elastic.

Dated the 28th day of March, 1946.

E. A. BINGEN,

Solicitor for the Applicants.

COMPLETE SPECIFICATION

Melt Blending of Aromatic Polyesters with other Polymeric Materials

85 We, JAMES TENNANT DICKSON AND REGINALD JOHN WILLIAM REYNOLDS, both of Hexagon House, Blackley, Manchester, British Subjects, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical
90 House, Millbank, London, S.W.1., a Company incorporated under the laws of Great Britain, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to the melt blending of aromatic polyesters with other polymeric materials and more par-

ticularly with other linear condensation polymers.

The aromatic polyesters with which the present invention is concerned are the highly polymeric linear esters obtainable by heating glycols of the series $\text{HO}(\text{CH}_2)_n\text{OH}$, where n is an integer greater than 1 but not exceeding 10, with terephthalic acid or with an ester-forming derivative thereof, for example, an aliphatic (including cycloaliphatic) or aryl ester or half-ester, an acid halide or an ammonium or amine salt, under conditions which yield the esters in a highly polymerised condition. Examples
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105
110

of such highly polymeric linear esters are those obtainable from terephthalic acid or an ester-forming derivative thereof and ethylene glycol, trimethylene glycol, tetramethylene glycol, hexamethylene glycol and decamethylene glycol. These polyesters are high melting, difficultly soluble, colourless or substantially colourless materials which can be formed into filaments which can be extended by drawing into strong flexible fibres showing, by characteristic X-ray patterns, molecular orientation along the fibre axis. Of these polyesters, polyethylene terephthalate is preferred on account of its ready availability and of its outstanding utility as a textile material.

We have now found that these aromatic polyesters may be satisfactorily blended by melting with a wide variety of other synthetic linear condensation polymers.

According to the present invention we provide blends of aromatic polyesters of the kind hereinbefore specified with other synthetic linear condensation polymers, said blends being obtained by heating said polyesters and the other polymers together above the melting points thereof at least until a homogeneous blend is obtained.

Synthetic linear condensation polymers which may be blended in accordance with the present invention include polyamides and interpolymides of all kinds, for example, polyhexamethylene adipamide, polyhexamethylene sebacamide, the polyamides obtainable from caprolactam, the interpolymides obtainable from caprolactam and hexamethylenediammonium adipate and the interpolymides from hexamethylenediammonium adipate, hexamethylenediammonium sebacate and caprolactam; polyesters and interpolymers (including different aromatic polyesters from the class hereinbefore specified), for example, polytrimethylene terephthalate, polyhexamethylene terephthalate, polydiethyleneglycol terephthalate, polyethylene-1:2-ethane bis(phenoxy-p-carboxylate), polyethylene adipate, polyethylene sebacate and polyethylene hexahydroterephthalate; polyurethanes and polyureas.

In carrying the invention into practical effect the components are mixed together and heated at a temperature at least sufficiently elevated to cause the components to melt, and the heating is continued until a homogeneous mass is obtained. Blending may be facilitated by stirring and if desired by the use of reduced pressures. Advantageously, the melting is effected in the absence of air

or oxygen and, if desired, a stream of an oxygen-free gas, for example, nitrogen, may be passed through the melt to assist in the formation of a homogeneous blend. If desired, the blending operation can be preceded by a drying operation.

The melts obtained at the conclusion of the heating operation may be extruded or otherwise removed from the vessel and subsequently cooled. The extruded melts may be formed into blocks, chips or other shapes.

The blends obtained according to the present invention usually find utility in the art in the form of filaments or fibres. Filaments can be formed by extrusion or by drawing from the melt directly after the completion of the heating or after re-melting. Any suitable apparatus may be used. The filaments can be subsequently drawn to as much as several times their original lengths. The drawing operation may be carried out on filaments which have been allowed fully to cool and solidify after their formation, or it may directly follow the formation of the filaments as one part of a continuous process. Any suitable apparatus and process may be used for the drawing. For instance, the filaments may be wound from one roller to another, the second roller rotating at a higher speed than the first roller, for example at a speed up to about 4 or 5 times that of the first roller.

In addition to their utility in the form of filaments and fibres the blends obtained according to the present invention may be formed from the melt into other shaped articles, for example, films or mouldings, or they may be used for hot melt coatings or as adhesives, plasticizers, binders for coating compositions or bonding materials for laminated fabrics, for example, in doubled fabric collars.

The invention is illustrated but not limited by the following Examples, in which the parts are expressed by weight.

EXAMPLE 1

18 parts of polyethylene terephthalate and 2 parts of polyhexamethylene adipamide, both in the form of small chips, are mixed together in a suitable vessel, dried by heating at 200°C. at a pressure of 15 mm. of mercury for 3 hours and heated at 275°C. in a current of oxygen-free nitrogen at the same pressure. The mass melts and becomes homogeneous after 3 hours heating. The blend is then extruded from the vessel in which it has been made and cooled. It is crystalline, has a melting point of 225°C.,

and it can be formed from the melt into filaments which can be extended by drawing into useful fibres.

EXAMPLE 2

5 Equal parts of polyethylene terephthalate and the interpolyamide formed from hexamethylenediammonium adipate, hexamethylenediammonium sebacate and caprolactam (40:30:30), both in the
10 form of small chips, are mixed together in a suitable vessel, which is fitted with a stirrer, and dried by heating for 3 hours at 200°C. at a pressure of about 15 mm. of mercury in a current of
15 oxygen-free nitrogen. The interpolyamide melts during this heating. The temperature is increased to 275°C. and the heating is continued otherwise under the same conditions. The polyethylene
20 terephthalate melts and after about 1½ hours a homogeneous blend is obtained. The blend is then extruded from the vessel and cooled. It is crystalline, has a melting point of 230°C., and it can be
25 formed from the melt into filaments which can be extended by drawing into useful fibres.

EXAMPLE 3

30 The recipe of Example 2 is repeated except that an equal part of polyethylene adipate is used in place of the interpolyamide. The blend obtained is crystalline, melts at 148°C., and it can be formed from the melt into filaments
35 which are somewhat elastic.

EXAMPLE 4

18 parts of polyethylene terephthalate and 2 parts of the polyurethane obtainable from tetramethylene glycol and
40 hexamethylene diisocyanate are mixed together in a suitable vessel, dried by heating at 198°C. at a pressure of 15 mm. of mercury for 3 hours in a current of oxygen-free nitrogen, and then heated
45 at 278°C. at the same pressure in a current of oxygen-free nitrogen. The mass melts and becomes homogeneous after 3 hours heating. The blend is then extruded from the vessel and cooled. It
50 is crystalline, has a melting point of 208°C., and it can be formed from the melt into filaments which can be extended by drawing into useful fibres.

EXAMPLE 5

55 Equal parts of polyethylene terephthalate and polyethylene-1,2-ethane bis (phenoxy - p - carboxylate) are mixed together in a suitable vessel, dried by
60 heating at 198°C. at a pressure of 15 mm. of mercury for 3 hours in a current of oxygen-free nitrogen, and then heated at 278°C. at the same pressure in a

current of oxygen-free nitrogen. The mass melts and becomes homogeneous after 3 hours heating. The blend is then
65 extruded from the vessel and cooled. It is crystalline, has a melting point of 170-180°C. and it can be formed from the melt into filaments, which can be extended by drawing into useful fibres. 70

EXAMPLE 6

Equal parts of polyethylene terephthalate and polyethylene hexahydroterephthalate are mixed together in a suitable
75 vessel, and the mixture is heated at 282°C. in a current of oxygen-free nitrogen for 2 hours at atmospheric pressure followed by two hours at a pressure of 0.5 mm. of mercury. The mixture which is initially heterogeneous
80 has become homogeneous at the end of the heating operation. The blend is extruded from the vessel in which it has been formed and cooled. A tough glass-like mass is obtained. The blend has a
85 melting point of 98-104°C., and it can be formed from the melt into filaments which are somewhat elastic.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:— 90

1. Blends of aromatic polyesters of the kind hereinbefore specified with other
95 synthetic linear condensation polymers, said blends being obtained by heating said polyesters and the other polymers together above the melting points thereof at least until a homogeneous blend is
100 obtained.

2. Blends as claimed in Claim 1 wherein there is used, as the aromatic polyester, polyethylene terephthalate.

3. Blends as claimed in either of the preceding Claims wherein the blending
105 is facilitated by stirring and, if desired, by the use of reduced pressures.

4. Blends as claimed in any of the preceding claims wherein melting is
110 effected in the absence of air or oxygen and, if desired, a stream of an oxygen-free gas, for example, nitrogen, is passed through the melt to assist in the formation of a homogeneous blend. 115

5. Blends as claimed in any of the preceding claims wherein the blending operation is preceded by a drying operation.

6. Blends of aromatic polyesters with other synthetic linear condensation polymers as hereinbefore particularly described and ascertained especially with
120 reference to the foregoing Examples.

Dated the 28th day of March, 1947.

E. A. BINGEN,

Solicitor for the Applicants.